Nonequilibrium Materials Engineering beyond Floquet

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Theoretical description of pump-probe spectroscopy in solids

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Driven is different

Kapitza pendulum

dynamical stabilization of a metastable state
Is driven also useful?

Exposing hidden states

Light-induced new states?

Possible light-induced superconductivity in $K_3C_{60}$ at high temperature

L Stojchevska et al. Science 2014;344:177-180

... and many more.
Pump-probe spectroscopy

- stroboscopic investigations of dynamic phenomena

Muybridge 1887

TbTe3 CDW metal

Image courtesy: J. Sobota / F. Schmitt
Grand Challenge #3: How do remarkable properties of matter emerge from complex correlations of the atomic or electronic constituents and how can we control these properties?

Grand Challenge #5: How do we characterize and control matter away – especially very far away – from equilibrium?
Challenge

movies by Koichiro Tanaka (Kyoto university)

many-body problem
(electrons + ions)

nonequilibrium many-body problem
(electrons + ions + photons)

Mission statement:
To understand and predict electron-ion dynamics and control of emergent nonequilibrium electronic structure
Challenge

Main challenges:
• hierarchy of energy and time scales
• high laser intensities: nonperturbative/nonlinear

Possible approaches:
• first principles (time-dependent density functional theory (TDDFT))
• effective models:
  – Feynman diagrams: self-energy
  – Keldysh nonequilibrium Green’s functions
  – connection with DFT: Sham-Schlüter integral equation
Ultrafast Materials Science today

**Understanding the nature of quasiparticles**
- Relaxation dynamics
- Control of couplings

PRX 3, 041033 (2013)  PRB 95, 205111 (2017)

**Understanding ordered phases**
- Collective oscillations
- Competing orders

PRB 93, 144506 (2016)  arXiv:1808.00712
arXiv:1810.06536

**Creating new states of matter**
- nonequilibrium topological states

Nature Comm. 6, 7047 (2015)
Nature Comm. 8, 13940 (2017)
Nature Comm. 9, 4452 (2018)

Image courtesy: D. Basov
Relaxation dynamics

**PRL 111, 077401 (2013)**
nonthermal pumped states

**PRB 87, 235139 (2013)**
extracting unoccupied electronic structure

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**PRX 3, 041033 (2013)**
small fluences

**PRB 90, 075126 (2014)**
fluence dependence

**Nat. Comm. 7, 13761 (2016)**
comparison with experiment
Electron-boson coupling

Holstein model (minimal version):

\[ H = \sum_k \epsilon(k) c_k^\dagger c_k + \Omega \sum_i b_i^\dagger b_i - g \sum_i c_i^\dagger c_i (b_i + b_i^\dagger) \]

Electrons (Fermi gas/liquid)  Bosons (e.g., Einstein phonon)  Electron-boson coupling

Pump laser:

\[ \epsilon(k) \rightarrow \epsilon(k, t) \]
Method: Keldysh Green functions

\[ G_k(\omega) = G^0_k(\omega) + G^0_k(\omega) \Sigma(\omega) G_k(\omega) \]

\[ G_k(t, t') = G^0_k(t, t') + \int dt_1 dt_2 G^0_k(t, t_1) \Sigma(t_1, t_2) G_k(t_2, t') \]

self-energy \( \Sigma \): electron-electron scattering electron-phonon scattering

...
Electron-boson coupling

Weak pump

Strong pump

t = -65.00

time unit = 0.66 fs

boson window effect for fast versus slow relaxation

nonlinear response for strong pump

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Orderer phases

**PRB 92, 224517 (2015)**

Higgs amplitude mode oscillations in pump-probe photoemission spectroscopy

**PRB 93, 144506 (2016)**

Light-enhanced superconductivity: electron-phonon scattering versus collective order parameter dynamics
Some recent key results

How to engineer materials away from equilibrium?

Part I: Light-enhanced electron-phonon coupling

Resonant excitation of IR phonon enhances electron-phonon coupling

E: Pomarico et al., PRB 95, 024304 (2017) – experiment (bilayer graphene)

M. A. Sentef, PRB 95, 205111 (2017) – theory

Part II: Optical control of chiral superconductors

Short laser pulses allow for switching of Majorana modes

M. Claassen et al., arXiv:1810.06536

Part III: From classical to quantized photon fields

Materials engineering in an optical cavity using vacuum fluctuations

M. A. Sentef et al., arXiv:1802.09437
I Resonant excitation of crystal lattice

M. Först et al., Nature Physics 7, 854 (2011)
Classical nonlinear phononics

Simplest model: classical dynamics

\[ \dot{Q}_{RS} + \Omega_{RS}^2 Q_{RS} = A Q_{IR}^2 \]

\[ \dot{Q}_{IR} + \Omega_{IR}^2 Q_{IR} = \frac{e^* E_0}{\sqrt{M_{IR}}} \sin(\Omega_{IR} t) F(t) \]

Rectified phonon field ➔ directional force

\[ H = A Q_{IR}^2 Q_{RS} \]

“nonlinear phononics“

M. Först et al., Nature Physics 7, 854 (2011)
Classical nonlinear phononics

Explains a number of observed effects, e.g.,

- structurally induced metal-insulator transitions
- phononic rectification in YBCO
  Mankowsky et al., Nature 516, 71 (2014)
- ferroelectric switching in LiNbO$_3$

**Classical phonon dynamics does not explain all effects in IR-driven materials.**

examples:  - light-induced superconductivity
              - light-enhanced el-ph coupling

... quantum nature of phonons important?
Enhanced electron-phonon coupling in graphene with periodically distorted lattice

E. Pomarico, M. Mitrano, H. Bromberger, M. A. Sentef, A. Al-Temimy, C. Coletti, A. Stöhr, S. Link, U. Starke, C. Cacho, R. Chapman, E. Springate, A. Cavalleri, and I. Gierz
Phys. Rev. B 95, 024304 – Published 13 January 2017

**PRB 95, 024304 (2017)**

enhanced electron-phonon coupling for pump on resonance with IR phonon
Dynamically enhanced coupling

PRB 95, 024304 (2017)

transient reduction of THz Drude weight
accelerated tr-ARPES relaxation

driving on phonon resonance: 3-fold enhancement of effective $\lambda_{el-ph}$
Quantum nonlinear phononics

2-site toy model, solve dynamics exactly

\[ \hat{H}(t) = -J \sum_{\sigma} (c_{1,\sigma}^\dagger c_{2,\sigma} + c_{2,\sigma}^\dagger c_{1,\sigma}) + g_2 \sum_{\sigma,l=1,2} \hat{n}_{l,\sigma} (b_{l} + b_{l}^\dagger)^2 \]

\[ + \Omega \sum_{l=1,2} b_{l}^\dagger b_{l} + F(t) \sum_{l=1,2} (b_{l} + b_{l}^\dagger), \]

Idea: Drive nonlinearly coupled IR-phonon, analyze electronic response

Drive: \[ F(t) = F \sin(\omega t), \]

Response: \[ I(\omega, t_0) = \Re \int dt_1 dt_2 e^{i\omega(t_1-t_2)} s_{t_1, t_2, \tau}(t_0) \]

time-resolved spectral function

\[ \times \left[ \langle \psi(t_2) | c_{1,\uparrow}^\dagger \mathcal{T} e^{-i \int_{t_1}^{t_2} H(t) dt} c_{1,\uparrow} | \psi(t_1) \rangle + + \langle \psi(t_1) | c_{1,\uparrow}^\dagger \mathcal{T} e^{-i \int_{t_2}^{t_1} H(t) dt} c_{1,\uparrow}^\dagger | \psi(t_2) \rangle \right], \]
IR-driven nonlinear el-ph system

Driving IR phonon with sinusoidal $F(t)$: coherent phonon oscillation

enhancement of local electronic double occupancy

$\rightarrow$ induced el-el attraction
Reduced coherence peaks with stronger driving

light-enhanced el-ph coupling

light-induced polaron formation

2-phonon shakeoff
Field dependence

Theory

Data by E. Pomarico, unpublished

Coherence peak weight loss: proportional to field intensity $F^2$ consistent with experiments

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Summary I

- enhanced electron-phonon coupling in phononically driven bilayer graphene

\[ PRB \ 95, \ 024304 \ (2017) \]

Exact solution of electron-phonon model system:

- theoretical proposal: nonlinear el-ph coupling as mechanism behind this enhancement

\[ PRB \ 95, \ 205111 \ (2017) \]
II Optical control of Majoranas

• prior work: optical control of competing orders

Theory of Laser-Controlled Competing Superconducting and Charge Orders

M. A. Sentef, A. Tokuno, A. Georges, and C. Kollath
Phys. Rev. Lett. 118, 087002 – Published 21 February 2017

– selective laser driving **switches** between phases
II Optical control of Majoranas

• can one switch the chirality of a 2D topological superconductor?

key idea: use two-pulse sequence with linearly and circularly polarized light
multiband Bogoliubov-de-Gennes Hamiltonians for doped graphene (d+id) and Sr2RuO4 (p+ip) coupling to fermionic reservoir to dissipate energy
laser driving via Peierls substitution

Keldysh equations of motion for Nambu Green’s functions:

\[ i\partial_t G_k(t, t') = \mathcal{H}_k(t, \Delta_k(t)) G_k(t, t') + \int d\tau \hat{\Sigma}_k(t, \tau) G_k(\tau, t') \]

\[ \Delta_k(t) = \frac{1}{L} \sum_j u^{(j)} \hat{\eta}_k^{(j)} \sum_{k', \alpha \beta} \hat{\eta}_{k' \alpha \beta} \left\langle \hat{c}_{-k', \beta \downarrow} \hat{c}_{k', \alpha \uparrow} \right\rangle \]
Optical control of Majoranas

A two-pulse sequence reverses d+id state in graphene.

Time-resolved spectroscopy tracks chirality reversal.
Summary II

• All-optical control of chiral Majorana modes
• towards arbitrarily programmable quantum computer?


M. Claassen
D. Kennes
III Cavity materials

• can one use enhanced vacuum fluctuations to change materials properties?
Cavity materials

**BCS superconductors:** phonon-mediated superconductivity

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Cavity-assisted mesoscopic transport of fermions: Coherent and dissipative dynamics.
Hagenmüller et al., 1801.09876

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Cavity-mediated electron-photon superconductivity
Frank Schlöwa, Andrea Cavalleri, and Dieter Jaksch

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Exciton-Polariton Mediated Superconductivity
Fabrice P. Laussy, Alexey V. Kavokin, and Ivan A. Shelykh

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Cavity Quantum Eliashberg Enhancement of Superconductivity
Jonathan B. Curtis, Zachary M. Raines, Andrew A. Allocco, Mohammad Hafezi, and Victor M. Galitski

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Manipulating quantum materials with quantum light
Martin Kiffner, Jonathan Coulthard, Frank Schlöwa, Arzhang Ardavan, and Dieter Jaksch

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Cavity superconductor-polaritons
Andrew A. Allocco, Zachary M. Raines, Jonathan B. Curtis, and Victor M. Galitski

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**Superradiant Quantum Materials**
Giacomo Mazza and Antoine Georges

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**Ab-initio Exciton-polaritons:** Cavity control of Dark Excitons in two dimensional Materials
Simone Latini, Enrico Ronca, Umberto De Giovannini, Hannes Hübscher, and Angel Rubio

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monolayer FeSe/STO

monolayer FeSe/STO: $T_c > 65$ K
bulk FeSe: $T_c = 9$ K

Huang and Hoffman, Annu. Rev. CMP 8, 311 (2017)
monolayer FeSe/STO: ARPES

replica bands: forward (small-q) electron-phonon scattering

monolayer FeSe/STO: interfacial phonon

bare el-phonon vertex

\[ g(q) = g_0 \exp\left(-|q|/q_0\right) \]

\[ q_0^{-1} = h_0 \sqrt{\epsilon_\parallel/\epsilon_\perp} \]

\[ \epsilon_\parallel/\epsilon_\perp \approx 100 \]

Lee et al., Nature 515, 245 (2014)

Huang and Hoffman, Annu. Rev. CMP 8, 311 (2017)
Cavity engineering

- idea: use **phonon polaritons** to enhance electron-phonon coupling

_Huang and Hoffman, Annu. Rev. CMP 8, 311 (2017)_
Model and Method

\[ H = \sum_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k}} c_{\mathbf{k}, \sigma}^\dagger c_{\mathbf{k}, \sigma} + \frac{1}{\sqrt{N}} \sum_{\mathbf{k}, \mathbf{q}, \sigma, \lambda = \pm} c_{\mathbf{k} + \mathbf{q}, \sigma}^\dagger c_{\mathbf{k}, \sigma} (g_\lambda^*(\mathbf{q}) \alpha_{\mathbf{q}, \lambda}^\dagger + g_\lambda(\mathbf{q}) \alpha_{\mathbf{q}, \lambda}) + \sum_{\mathbf{q}, \lambda = \pm} \omega_\lambda(\mathbf{q}) \alpha_{\mathbf{q}, \lambda}^\dagger \alpha_{\mathbf{q}, \lambda} \]

- electrons
- el-polariton coupling
- polaritons

bare el-phonon vertex

G-self-consistent Migdal-Eliashberg diagram

\[ g(\mathbf{q}) = g_0 \exp(-|\mathbf{q}|/q_0) \quad q_0^{-1} = h_0 \sqrt{\epsilon_\parallel/\epsilon_\perp} \]

Mass enhancement: \( m^*/m = 1 + \lambda \)
Cavity materials: Phonon polaritons

\[ \omega(q) = \omega_+ (\text{upper polariton}) + \Omega (\text{phonon}) + \omega_- (\text{lower polariton}) \]

Enhanced electron-phonon coupling, controlled by cavity volume
Superconductivity

suppressed superconductivity despite enhanced el-ph coupling

\[ T_C \approx \frac{\lambda \Omega}{2 + 3\lambda} \]

\( T_{C, BCS} \approx 1.13\Omega \exp\left(-\frac{1}{\lambda}\right) \)

forward scattering vs. q-independent scattering
Summary III

- cavity leads to **enhanced electron-phonon coupling**
- can one also enhance superconductivity?

Summary

Ultrafast laser engineering of

- band structure, topology (Floquet)
  
  * Nature Commun. 6, 7047 (2015)
  * Nature Commun. 8, 13940 (2017)
  * arXiv:1803.07447

- electron-phonon coupling
  
  * PRB 95, 024304 (2017)
  * PRB 95, 205111 (2017)
  * arXiv:1802.09437

- Hubbard model with strong subresonant band structure, topology (Floquet)
  
  * PRL 121, 097402 (2018)

- ordered phases
  
  * PRB 93, 144506 (2016) arXiv:1808.00712
  * arXiv:1810.06536

Towards nonequilibrium materials engineering
Outlook: Group projects

R. Tuovinen (postdoc): nonequilibrium Green’s functions (GKBA) for time-resolved transport and excitonic condensates (JCTC 14, 2495 (2018); arXiv:1808.00712)


S. Ramirez (PhD student) light-induced Majoranas

M. Kalthoff (PhD student) time-dependent matrix product states (t-DMRG) for Floquet engineering of correlated systems (w/ D. Kennes, FU Berlin)

D. Hofmann (master student) topological exciton polaritons (master), machine learning for time-dependent variational wave functions (w/ G. Carleo, CCQ NYC)

X. Wang (student, Tsinghua) Green’s functions for cavity 2D materials with focus on topology

M. Puviani (PhD st., Modena) quantum nonlinear phononics, ultrafast melting of ferrielectric charge-density wave (arXiv:1806.08187, PRB)